# Probing two-particle orbital orientation of bound states 

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received 16 November 2004; accepted in final form 4 February 2005
published online 9 March 2005
PACS. 32.80. Fb - Photoionization of atoms and ions.
PACS. 31.25.Jf - Electron correlation calculations for atoms and ions: excited states.
PACS. 74.20.-z - Theories and models of superconducting state.


#### Abstract

Under certain conditions specified in this work, a two-particle orbital orientation of bound states can be detected by means of double photoemission with polarized light. These conclusions are based on a tensorial analysis of the dipole transition amplitudes and are illustrated by a numerical study of double photoemission from helium oriented by laser-pumping. The analytical and numerical results demonstrate the interrelation between two-particle correlation and orbital polarization.


Introduction. - In most metals low-energy electrons can be described by the Landau theory of Fermi liquids. The elementary excitations of such a liquid are quasiparticles that move almost independently and with a well-defined energy-momentum dispersion relation [1]. At low temperatures $T$, however, many of these liquids become unstable due to inter-quasiparticle interactions resulting in a spontaneous symmetry breaking and in the formation of a new state of matter, such as ferromagnetism and superconductivity (SC) [2,3]. Cooper instability in conventional SC leads to the formation of $s$-wave Cooper pairs with a vanishing total spin. In this case gauge symmetry is broken at the critical temperature $T_{\mathrm{c}}$ [3]. On the other hand, various experimental and theoretical studies have established new states of unconventional SC associated with other kind of spontaneous symmetry breaking. For instance, in case of triplet $p$-wave SC, which is believed to occur in the layered $\mathrm{Sr}_{2} \mathrm{RuO}_{4}$ compound [3], for $T \leq T_{\mathrm{c}}$ the time-reversal symmetry is broken in the superconducting ground state (which is also called the chiral or the Anderson-Brinkmann-Morel (ABM) phase [2,3]). In view of this established knowledge and numerous other examples it is valuable to envision methods and ways that test for inter-particle correlations as well as for the footprints of possible symmetry reduction. A recent work [4] along this line has demonstrated that correlation within a Cooper pair in a conventional $s$-wave SC can be mapped out by means of one-photon two-electron spectroscopy (also called double photoemission DPE [5]). The question we pose here is whether and under which conditions it is possible to test by means of DPE for a break of time-reversal symmetry in a given sample and to test at the same time for the influence of quasi-particle correlation. Surprisingly, this question is still unaddressed even for the most "simple" case of a two-electron atom or ion. The present work is an attempt to deliver an answer to the

[^0]above problem in a general manner and without relying on special properties akin to a specific system. The approach is based on a tensorial analysis of the DPE transition amplitude from an arbitrary two-electron state oriented along a given direction $\hat{\boldsymbol{a}}$. The key conclusions are: The difference in DPE cross-sections for two different helicity state of a circularly polarized photon (this difference is called conventionally circular dichroism, CD) signifies, under certain experimental arrangements specified below, the existence of a well-defined dynamical phase $(\phi)$ of the two-particle wave function. The influence of reversing the sign of this phase is measurable and is equal to CD. (Quasi)particle correlation is reflected in a dependence of the CD (and hence of $\phi$ ) on the inter-particle quantities, such as the relative momenta and energies. For time-reversal symmetric states CD vanishes. We remark that CD is as well present in single photoemission processes and can be employed to test for spontaneous breaking of time-reversal symmetry (of single-particle orbitals), as demonstrated experimentally for the pseudogap state of high- $T_{\mathrm{c}}$ SC [6]. DPE gives access to the properties of electron pairs and offers additional information in that it allows an insight into the interplay between the symmetry reduction and the inter-particle correlation. This is particularly important for the understanding of the underlying mechanism of high- $T_{\mathrm{c}}$ SC.

To illustrate our findings and to give an estimate of the value of CD we performed fullnumerically the first calculations for DPE from oriented two-particle states, namely from $2^{3} P_{m= \pm 1}$ and $2^{1} P_{m= \pm 1}$ of helium. Here the chirality of the initial electron pair state is supposed to be created experimentally (in contrast to the ABM ground state), for example, by laser pumping ortho- or para-helium ground state with circularly polarized light. A markedly different CD is obtained for triplet and singlet states which illustrates the influence of the exchange interaction on $\phi$.

General formulation. - We consider a target (orbitally) oriented along $\hat{\boldsymbol{a}}$ and described by an effective correlated two-particle state $\chi_{12} \varphi_{\hat{\boldsymbol{a}}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$, where $\chi_{12}$ is the spin part and $\varphi_{\hat{\boldsymbol{a}}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$ is the spatial part of the wave function with a certain parity $\left({ }^{1}\right)$. By definition, the oriented (chiral) state has a non-zero state multipole of the first rank (proportional to the mean value of the angular-momentum vector) and therefore for such a state the time-reversal symmetry is broken. We focus on systems in which spin and spatial degrees of freedom are decoupled, i.e. the spin part $\chi_{12}$ dictates only the symmetry of $\varphi_{\hat{a}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$ under particle exchange. A circularly polarized photon with a polarization vector $\hat{\boldsymbol{\epsilon}}_{+}\left(\hat{\boldsymbol{\epsilon}}_{-}\right)\left({ }^{2}\right)$ excites this initial state to a scattering state $\chi_{12}^{\prime} \psi_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$ which is characterized by the wave vectors $\boldsymbol{k}_{1}, \boldsymbol{k}_{2}$ of the emitted electrons. In the dipole approximation used below the photon does not couple to the electron spins. Thus, $\chi_{12}^{\prime}=\chi_{12}$ and the optical transition amplitude has the form

$$
\begin{equation*}
T_{\hat{\boldsymbol{\epsilon}}_{ \pm}}=\hat{\boldsymbol{\epsilon}}_{ \pm} \cdot \boldsymbol{f}_{\hat{\boldsymbol{a}}}, \quad \boldsymbol{f}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)=\left\langle\psi_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}\right| \boldsymbol{D}\left|\varphi_{\hat{\boldsymbol{a}}}\right\rangle, \tag{1}
\end{equation*}
$$

where $\boldsymbol{D}$ is the dipole operator. The DPE cross-sections $\sigma$ have the structure $\sigma\left(\hat{\boldsymbol{\epsilon}}_{ \pm}\right)=c\left|T_{\hat{\boldsymbol{\epsilon}}_{ \pm}}\right|^{2}$ with $c$ being a real factor. From eq. (1) it follows that

$$
\begin{equation*}
\left|T_{\hat{\epsilon}_{ \pm}}\right|^{2}=\left(\hat{\epsilon}_{ \pm} \cdot \boldsymbol{f}_{\hat{\boldsymbol{a}}}\right)\left(\hat{\epsilon}_{ \pm}^{\hat{*}} \cdot \boldsymbol{f}_{\hat{\boldsymbol{a}}}^{*}\right), \tag{2}
\end{equation*}
$$

thus

$$
\begin{equation*}
\left|T_{\hat{\epsilon}_{ \pm}}\right|^{2}=\frac{1}{3}\left|\boldsymbol{f}_{\hat{\boldsymbol{a}}}\right|^{2}+\frac{1}{2}\left(\hat{\boldsymbol{\epsilon}}_{ \pm} \times \hat{\boldsymbol{\epsilon}}_{ \pm}^{*}\right) \cdot\left(\boldsymbol{f}_{\hat{\boldsymbol{a}}} \times \boldsymbol{f}_{\hat{\boldsymbol{a}}}^{*}\right)+\mathrm{T}_{2}\left(\hat{\boldsymbol{\epsilon}}_{ \pm}, \hat{\boldsymbol{\epsilon}}_{ \pm}^{*}\right) \mathrm{T}_{2}\left(\boldsymbol{f}_{\hat{\boldsymbol{a}}}, \boldsymbol{f}_{\hat{\boldsymbol{a}}}^{*}\right) . \tag{3}
\end{equation*}
$$

[^1]$\left({ }^{2}\right)$ We use a convention that a right (left) circularly polarized light has $\hat{\boldsymbol{\epsilon}}_{+}\left(\hat{\boldsymbol{\epsilon}}_{-}\right)$polarization vector.

Here $\mathrm{T}_{2}(\boldsymbol{x}, \boldsymbol{y})$ is a spherical tensor of rank 2 constructed from the spherical components of the vectors $\boldsymbol{x}$ and $\boldsymbol{y}$ [9]. Only the second term on the rhs of eq. (3) is odd with respect to exchange of $\hat{\boldsymbol{\epsilon}}_{+}$and $\hat{\boldsymbol{\epsilon}}_{-}$(or $\boldsymbol{f}_{\hat{\boldsymbol{a}}}$ and $\boldsymbol{f}_{\hat{\boldsymbol{a}}}^{*}$ ). Therefore, the measurable circular dichroism, defined as $\mathrm{CD}=\sigma\left(\hat{\boldsymbol{\epsilon}}_{+}\right)-\sigma\left(\hat{\boldsymbol{\epsilon}}_{-}\right)$, reads

$$
\begin{equation*}
\mathrm{CD}=c\left[\left|T_{\hat{\boldsymbol{\epsilon}}_{+}}\right|^{2}-\left|T_{\hat{\boldsymbol{\epsilon}}_{-}}\right|^{2}\right]=-i c \hat{\boldsymbol{k}} \cdot\left(\boldsymbol{f}_{\hat{\boldsymbol{a}}} \times \boldsymbol{f}_{\hat{\boldsymbol{a}}}^{*}\right), \tag{4}
\end{equation*}
$$

where $\hat{\boldsymbol{k}}= \pm i \hat{\boldsymbol{\epsilon}}_{ \pm} \times \hat{\boldsymbol{\epsilon}}_{ \pm}^{*}$ is a real unit vector along the light propagation direction. The vector product $f_{\hat{a}} \times f_{\hat{a}}^{*}$ is pure imaginary, for $f_{\hat{a}} \times f_{\hat{a}}^{*}=-\left(f_{\hat{a}} \times f_{\hat{a}}^{*}\right)^{*}$, therefore dichroic effects disappear if $\boldsymbol{f}_{\hat{\boldsymbol{a}}}$ is pure real or pure imaginary.

Initial-state orientation. - To illustrate the physical significance of the above analysis we assume at first that the final state is treated within the plane-wave approximation (PWA) $\left({ }^{3}\right)$. It is known that within the PWA for isotropic initial state CD vanishes identically [10]. Therefore, a non-zero CD is an indicator for the oriented initial state. Applying the dipole approximation in the length form for $\boldsymbol{D}$, the vectors $\boldsymbol{f}_{\hat{\boldsymbol{a}}}$ (eq. (1)) acquire the form

$$
\begin{align*}
\boldsymbol{f}_{\hat{\boldsymbol{a}}} & =\frac{i}{(2 \pi)^{3}} \boldsymbol{\nabla} \int \mathrm{~d}^{3} \boldsymbol{r}_{1} \mathrm{~d}^{3} \boldsymbol{r}_{2} e^{-i \boldsymbol{k}_{1} \cdot \boldsymbol{r}_{1}-i \boldsymbol{k}_{2} \cdot \boldsymbol{r}_{\mathbf{2}}} \varphi_{\hat{\boldsymbol{a}}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right), \\
& =i \boldsymbol{\nabla} \tilde{\varphi}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right) \tag{5}
\end{align*}
$$

where $\boldsymbol{\nabla}=\boldsymbol{\nabla}_{\boldsymbol{k}_{1}}+\boldsymbol{\nabla}_{\boldsymbol{k}_{2}}$ and $\tilde{\varphi}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)$ is the electron pair momentum-space wave function. From eq. (4) there follows

$$
\begin{equation*}
\mathrm{CD}=-i c \hat{\boldsymbol{k}} \cdot\left[\boldsymbol{\nabla} \tilde{\varphi}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right) \times \boldsymbol{\nabla} \tilde{\varphi}_{\hat{\boldsymbol{a}}}^{*}\left(-\boldsymbol{k}_{1},-\boldsymbol{k}_{2}\right)\right] . \tag{6}
\end{equation*}
$$

Now it is evident that CD changes sign upon time-reversal operation $\left({ }^{4}\right)$ acting on the initial bound state and vanishes when the initial state is time-reversal invariant.

Reversing the sign of the phase of the spacial initial bound state corresponds to reversing the sign of the momentum-space wave function phase $\phi\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)$ and to inverting the momenta at the origin, i.e. $\varphi_{\hat{a}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right) \mapsto \varphi_{\hat{\boldsymbol{a}}}^{*}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$ corresponds to $\tilde{\varphi}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right) \mapsto \tilde{\varphi}_{\hat{\boldsymbol{a}}}^{*}\left(-\boldsymbol{k}_{1},-\boldsymbol{k}_{2}\right)$. Therefore, eq. (6) dictates $\mathrm{CD}=\sigma\left(\hat{\boldsymbol{\epsilon}}_{+}, \phi\right)-\sigma\left(\hat{\boldsymbol{\epsilon}}_{-}, \phi\right)=-\sigma\left(\hat{\boldsymbol{\epsilon}}_{+},-\phi\right)+\sigma\left(\hat{\boldsymbol{\epsilon}}_{-},-\phi\right)$. Comparing eqs. (4), (5) we also conclude that

$$
\begin{align*}
\mathrm{CD} & =\sigma\left(\hat{\boldsymbol{\epsilon}}_{+}, \phi\right)-\sigma\left(\hat{\boldsymbol{\epsilon}}_{-}, \phi\right)=\sigma\left(\hat{\boldsymbol{\epsilon}}_{+}, \phi\right)-\sigma\left(\hat{\boldsymbol{\epsilon}}_{+},-\phi\right), \\
& =\sigma\left(\hat{\boldsymbol{\epsilon}}_{-},-\phi\right)-\sigma\left(\hat{\epsilon}_{-}, \phi\right) . \tag{7}
\end{align*}
$$

The relations (7) allow the following key conclusions: The circular dichroism can be regarded as a measurable change in the cross-section upon the phase sign reversal of the initial twoelectron bound-state wave function. For real initial-state wave functions (e.g., if the state is isotropic) $\phi \equiv 0$ and hence $\mathrm{CD}=0$. A direct relation between the circular dichroism and the phase within PWA can be obtained from eqs. (4), (5):

$$
\begin{equation*}
\mathrm{CD}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)=\boldsymbol{C} \cdot \boldsymbol{\nabla} \phi\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right) ; \quad \boldsymbol{C}=c\left\{\left(\boldsymbol{\nabla}\left|\tilde{\varphi}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)\right|^{2}\right) \times \hat{\boldsymbol{k}}\right\} . \tag{8}
\end{equation*}
$$

[^2]As pointed out in ref. [11], the two-particle probability density $\left|\tilde{\varphi}_{\hat{\boldsymbol{a}}}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)\right|^{2}$ is accessed (within PWA) by using linear polarized light; hence for given $\boldsymbol{k}_{1}, \boldsymbol{k}_{2}$, one performs the experiments with linear and circular polarized light and retrieves the phase gradient from CD and the DPE cross-section $\left({ }^{5}\right)$.

Effects of final-state phase. - The direct connection between the initial-state phase and CD relies on the PWA, in which case additional dynamical phases due to distortions of the final-state wave function are eliminated. In general, however, the vector $\boldsymbol{f}_{\hat{a}}$, eq. (1), may be expressed as

$$
\begin{equation*}
\boldsymbol{f}_{\hat{\boldsymbol{a}}}=i \int \mathrm{~d}^{3} \boldsymbol{q}_{1} \mathrm{~d}^{3} \boldsymbol{q}_{2} \tilde{\psi}_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}^{*}\left(\boldsymbol{q}_{1}, \boldsymbol{q}_{2}\right) \boldsymbol{\nabla} \tilde{\varphi}_{\hat{a}}\left(\boldsymbol{q}_{1}, \boldsymbol{q}_{2}\right) \tag{9}
\end{equation*}
$$

where $\tilde{\psi}_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}\left(\boldsymbol{q}_{1}, \boldsymbol{q}_{2}\right)$ is the Fourier transform of $\psi_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$. Thus the phase of $\boldsymbol{f}_{\hat{\boldsymbol{a}}}$ is determined by the phases of $\tilde{\psi}_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}\left(\boldsymbol{q}_{1}, \boldsymbol{q}_{2}\right)$ and $\tilde{\varphi}_{\hat{a}}\left(\boldsymbol{q}_{1}, \boldsymbol{q}_{2}\right)$. These two-phase relations can be, however, disentangled by means of their symmetry properties, even in the general case. This is achieved by performing the experiment under specified geometrical conditions in which case the link between the dichroism and initial-state phase differences is generally established, i.e. it persists even in the presence of final-state interaction. To show that we recall that theory [10] and experiment [13] proved if the initial state is isotropic CD is finite $\left({ }^{6}\right)$, unless $\boldsymbol{k}_{1}, \boldsymbol{k}_{2}$ and $\boldsymbol{k}$ are coplanar or $k_{1}=k_{2}$. These conditions for vanishing CD for isotropic initial states we call non-chiral kinematics. In general, under these conditions a reversal of $\hat{\boldsymbol{k}}$ to $-\hat{\boldsymbol{k}}$ (or equivalently $\hat{\boldsymbol{\epsilon}}_{+}$to $\hat{\boldsymbol{\epsilon}}_{-}$) does not affect $\sigma$, unless there exists an additional polarization direction $\hat{\boldsymbol{a}}$ for the initial state. Thus, CD measurements unravel the orbital polarization, i.e. an orientation or an alignment of the state $\varphi_{\hat{\boldsymbol{a}}}$ along $\hat{\boldsymbol{a}}$ [14]. Measuring in the non-chiral kinematics a finite CD from a certain specimen does not determine whether the sample quantum state is aligned (along an axis) or oriented (along a direction). However, when the polarization direction is known from measurements $\left({ }^{7}\right)$ or from symmetry considerations and $\hat{\boldsymbol{k}} \| \hat{\boldsymbol{a}}$ one can choose a special DPE kinematics where one of the electrons is emitted along the quantization axis (cf. figs. 1). In this situation, the alignment does not contribute to $\mathrm{CD}[8]$ and CD is finite only if the initial state is oriented, as demonstrated numerically below.

Numerical examples. - To illustrate the above findings and to give an estimate of CD for real systems we calculated fully differential DPE cross-sections for orbitally polarized helium. The ground state of He has $J_{0}=0$ and cannot be polarized. However, the states $2^{1,3} P$ can be oriented, e.g. by pumping with a circularly polarized laser. Experimentally the use of the excited He as a target is quite feasible, as was demonstrated by studies on the superelastic electron scattering from laser excited $2^{3} P$ He state [15].

For clarity the calculations are performed using a simple (but reasonable) ground state wave functions which consist of (anti)symmetrized product of two hydrogen-like single-electron orbitals $u_{n l}$ with effective charges considered as variational parameters $\left({ }^{8}\right)$ :

$$
\begin{equation*}
\varphi_{m}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)=N_{ \pm}\left[u_{1 s}\left(r_{1}\right) u_{2 p}\left(r_{2}\right) Y_{1 m}\left(\hat{\boldsymbol{r}}_{2}\right) \pm u_{1 s}\left(r_{2}\right) u_{2 p}\left(r_{1}\right) Y_{1 m}\left(\hat{\boldsymbol{r}}_{1}\right)\right], \tag{10}
\end{equation*}
$$

[^3]

Fig. $1-m$-resolved double ionization cross-section of $2^{1} P_{m}$ state of atomic helium following the absorption of a right (left) circularly polarized photon, labelled respectively by $\epsilon_{+}\left(\epsilon_{-}\right)$. One slow electron with 5 eV energy is fixed along the light propagation direction $\boldsymbol{k}$ (which is also the quantization axis of the atom (thick arrow)). The polar angle $\theta$ of the second 10 eV electron is varied. The calculations were done using the 3C model for left (a) and right (b) circularly polarized light and within PWA (c) for right circularly polarized light. (d) the same as in (a), but for the triplet state $2^{3} P_{m}$. The results for $\hat{\epsilon}_{+}$can be obtained from those shown in the figure applying eq. (11).
where $N_{ \pm}$is a normalization factor and $Y_{1 m}(\hat{\boldsymbol{r}})$ are the spherical harmonics. The $+(-)$ sign refers to the singlet $2^{1} P$ (triplet $\left.2^{3} P\right)$ state. The electron-pair states with $m= \pm 1$ are completely oriented along $\pm z$-direction. Taking into account that $Y_{1 m}(\hat{\boldsymbol{r}})=(-1)^{m} Y_{1-m}^{*}(\hat{\boldsymbol{r}})$ one can show that in the chosen non-chiral kinematics the relation $\left(\hat{\boldsymbol{\epsilon}}_{+} \cdot \boldsymbol{f}_{m}\right)=\left(\hat{\boldsymbol{\epsilon}}_{-} \cdot \boldsymbol{f}_{-m}\right)^{*}$ holds and therefore

$$
\begin{equation*}
\sigma\left(\hat{\boldsymbol{\epsilon}}_{+}, m\right)=\sigma\left(\hat{\boldsymbol{\epsilon}}_{-},-m\right) \tag{11}
\end{equation*}
$$

Thus equations of the type (7) proved for the PWA are valid in a more general case. The final state $\psi_{\boldsymbol{k}_{1}, \boldsymbol{k}_{2}}$ is approximated by a product of two plane waves modified by three twobody Coulomb distorting factors, the so-called 3C approximation [17]. The 3C wave function accounts correctly for all two-particle multiple scattering processes (and hence it is asymptotically correct) but it neglects short-range correlations such as three-body scattering events. Nevertheless, for the purpose of illuminating the main physical effects (which is our aim here) the 3C model turned out to be very instructive and in most cases sufficient (for a detailed discussion of the strengths and shortcomings of the 3C theory we refer to ref. [5]). The functions $f_{\hat{a}}$ are evaluated full-numerically.

Figures 1(a) and 1(b) illustrate the message of this work. Dichroism caused by finalstate phase effects is eliminated by tuning the experimental arrangements to the non-chiral kinematics with one of the electrons moving along the $z$-axis. Then, DPE from a state with a time-reversal symmetry (aligned or isotropic state), such as $\varphi_{1,3 P_{m=0}}=\varphi_{1,3}^{*} P_{m=0}$, does not
show CD. On the other hand, the existence of a defined positive or negative sense of current circulation (around the directed vector $\hat{\boldsymbol{k}}$ ) associated with the electron pair state results in a finite CD. It suffices to measure DPE and CD from one orbitally oriented state to know the results for the (unknown) state with the opposite phase sign, as stated by eqs. (7, 11) and confirmed by calculations (cf. figs. 1(a) and 1(b)). This finding is decisive to test for orbital orientation of a given sample. It is important to recall that the phase $\phi$ of interest here is that of the correlated electron pair. Thus, the phase $\phi$ will depend on the initial-state correlation between the two electrons. Figures 1(a) and (d) demonstrate this dependence on one type of correlation, namely the exchange interaction. As clear from eq. (10), the initial state is the sum or difference of two (non-identical) complex functions, each having its own phase. Therefore, the phase $\phi_{+}$of the sum (singlet state) will be generally different from the phase $\phi_{-}$of the difference (triplet state), as demonstrated by figs. 1(a) and (d). An explicit analytic formula for the connection between the phase and CD is established only within PWA through eq. (8). When more realistic final-state wave functions (such as 3C) are used no such explicit connection has yet been established. Even though the existence of a phase of $\boldsymbol{f}_{\hat{a}}$ is exclusively due to the initial state (under the non-chiral kinematics), the value of the phase of $\boldsymbol{f}_{\hat{a}}$ remains obscured. This is because $\boldsymbol{f}_{\hat{\boldsymbol{a}}}$ is expressed (eq. (9)) as the integral of complex functions. For this reason, generally, a finite CD is an indicator for the existence of broken time-reversal symmetry in a system. The dependence of CD on $\boldsymbol{k}_{1}$ and $\boldsymbol{k}_{2}$ reveals the interplay between the time-reversal symmetry breaking and the inter-particle correlation.

As clear from fig. 1 the time-reversal symmetry is restored at certain configurations $\boldsymbol{k}_{1}$ and $\boldsymbol{k}_{2}$ (points where $\sigma(m=+1)=\sigma(m=-1)$ ), which within the PWA can be related to a vanishing (or constant) phase of the momentum-space two-electron wave function (cf. eq. (8)). These high symmetry points can be shifted (or removed) by final-state interaction (cf. figs. 1(b) and 1 (c) and eq. (9) for the underlying reason). With increasing energies $E_{1}$ and $E_{2}$ the PWA results become more reliable. In fact, figs. 1(b) and 1(c) evidence that, even at low energies, the PWA captures the overall features of the process, except when the two electrons are emitted in the forward hemisphere close to each other. In this case the electron-electron repulsion, which is not included in the PWA, is dominant and leads to a reduction of the cross-section in these regions. The angular correlation pattern originate from an interplay between dipole selection rules, exchange interaction and electron-electron electrostatic repulsion [8].

Final remarks and conclusions. - In summary we have shown that under certain conditions DPE can be utilized to study the existence of two-particle orbital orientation in correlated systems and it allows to map out the phase dependence on the two-particle correlation. This is demonstrated explicitly for the case of a polarized atomic helium target. As mentioned in the introduction we expect the proposed method to be useful for addressing the phase relation in superconductive materials such as those studied recently [18]. For this case in particular and for extended systems in general screening effects validate the use of the plane-wave approximation for the final state. This statement has been confirmed by a recent experiment [19]. However, when dealing with SC an additional complication arises: For high-temperature superconductors with a pairing mechanism the coherence length, i.e. the extent of the Cooper pair, is on the order of few lattice constants and the dipole approximation (on which the above theory is based) can be used. For conventional superconductors however the coherence length is on the micrometer scale and one has to deal with all the multipoles in the photon-target interaction. In this case one can also show [20] that the dichroism is related to the gradient of the phase of the Cooper pair wave function. In ref. [20] concrete calculations can be found for copper oxide high- $T_{\mathrm{c}}$ materials with a $d_{x^{2}-y^{2}}$ wave pairing symmetry in the $\mathrm{CuO}_{2}$ planes. The

SC gap is then real and hence CD vanishes identically. However, as proposed in refs. [21,22], upon an admixture of $s$-wave pairing the SC gap turns complex and the CD is then finite, as explicitly shown in ref. [20] where other types of pairing with time-reversal symmetry breaking are analyzed.

We thank A. Grum-Grzhimailo and K. A. Kouzakov for useful discussions and comments. NMK acknowledges the hospitality and the financial support by the MPI für Mikrostrukturphysik, Halle.

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[^1]:    $\left({ }^{1}\right)$ Many-particle systems can be mapped onto an effective two-particle problem [7]. The theory presented here is then valid within the frozen core approximation, i.e. if only the two-particle orbital is excited which has absorbed the photon. A more general approach based on the density matrix formalism can be found elsewhere [8].

[^2]:    $\left({ }^{3}\right)$ The plane-wave approximation (PWA) is unrealistic for atomic targets unless the electron energies are high compared to the initial binding. For solids screening effects make PWA more viable. In a superconductor (SC) the coherence length (the size of the pair) can reach a micron in conventional SC or few lattice constants in high- $T_{\mathrm{c}}$ SCs making the screening even more effective. This fact is endorsed by the agreement of the recent numerical PWA results with experimental data for single photoemission from conventional SC, cf. ref. [4] and references therein.
    $\left({ }^{4}\right)$ Under a time-reversal operation $\Theta$ the transformation applies $\Theta \varphi\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)=\varphi^{*}\left(\boldsymbol{r}_{1}, \boldsymbol{r}_{2}\right)$ and $\Theta \tilde{\varphi}\left(\boldsymbol{k}_{1}, \boldsymbol{k}_{2}\right)=$ $\tilde{\varphi}^{*}\left(-\boldsymbol{k}_{1},-\boldsymbol{k}_{2}\right)$.

[^3]:    $\left.{ }^{5}\right)$ In fact, the DPE cross-section with linear polarized light is related to $\sigma\left(\hat{\boldsymbol{\epsilon}}_{+}\right)+\sigma\left(\hat{\boldsymbol{\epsilon}}_{-}\right)$[12].
    ${ }^{(6)}$ )For isotropic targets the CD in DPE can be related to the dynamical phase of the final-state wave function [10].
    $\left({ }^{7}\right)$ The DPE cross-section $\sigma$ possesses a cylindrical symmetry around $\hat{\boldsymbol{k}}$ if $\hat{\boldsymbol{k}} \| \hat{\boldsymbol{a}}$. This fact can be used, in principle, to determine the angle between $\hat{\boldsymbol{a}}$ and $\hat{\boldsymbol{k}}$.
    $\left.{ }^{8}\right)$ For the $2^{1} P$ state the values of the variational parameters are $Z_{1 s}=2$ and $Z_{2 p}=0.97$ [16] which gives a binding energy of $E\left(2^{1} P\right)=-2.123$ (experimental value is -2.124 ). For the $2^{3} P$ state one finds $Z_{1 s}=1.99$ and $Z_{2 p}=1.09$ [16] leading to the binding energy of $E\left(2^{3} P\right)=-2.131$ (experimental value is -2.133 ).

