Generalized Stern-Gerlach Effect for Chiral Molecules

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(Received 4 May 2007; published 26 September 2007)

The Stern-Gerlach effect is well known as spin-dependent splitting of a beam of atoms with magnetic moments by a magnetic-field gradient. Here, we show that an induced gauge potential may lead to a similar effect for chiral molecules. In the presence of three inhomogeneous light fields, the center of mass of a three-level chiral molecule is subject to an optically induced gauge potential, and the internal dynamics of the molecule can be described as an adiabatic evolution in the reduced pseudospin subspace of the two lowest energy levels. We demonstrate numerically that such an induced gauge potential can lead to observable pseudospin-dependent and chirality-dependent generalized Stern-Gerlach effects for mixed left- and right-handed chiral molecules under realistic conditions.

DOI: 10.1103/PhysRevLett.99.130403 PACS numbers: 03.65.Vf, 11.15.−q, 42.50.−p

Introduction.—The Stern-Gerlach experiment [1] is one of the milestones in the development of quantum theory. The observation of the splitting of a beam of silver atoms in their ground states in a nonuniform magnetic field led to the concept of the electronic spin. In this Letter we show that even in the absence of a magnetic-field gradient, the center of mass of certain atoms or molecules in optical fields follows different trajectories corresponding to different inner states. This phenomenon is straightforwardly explained as a generalized Stern-Gerlach effect in the optically induced gauge potential [2–4].

This induced gauge potential consists of effective vector and scalar potentials, which result from the adiabatic variable separation of the slow spatial and fast inner dynamics of the atom according to the generalized Born-Oppenheimer approximation [4]. Recently, there has been considerable interest in the implementation of various pseudospin-dependent induced gauge potentials for cold atoms. Examples include the induced monopole [5], and the spin Hall effect for cold atoms [6,7], in direct analogy to the spin Hall effect due to the spin-orbit coupling in condensed matter physics [8]. Here, we would like to consider consequences of the induced gauge potential in systems of cold chiral molecules [9–11] that manifest themselves as a generalized Stern-Gerlach effect.

We consider a chiral molecule (see Fig. 1), which is described by a cyclic three-level system [9,10,12–14] where any two of the three levels are coupled by a classical optical field. A specific example is cyclic three-level (Δ-type) chiral molecules, e.g., the D_2S_2 enantiomers in Ref. [10] when only the lowest three states in each well are considered. Such symmetry-breaking systems can also be implemented using an asymmetric well and its mirror [9] (i.e., one asymmetric well and its mirror form a symmetric double well which supports chirality), or a superconducting circuit acting as an effective atom [12]. It will be shown that the optically induced gauge potentials for the chiral molecules will be both chirality dependent and pseudospin dependent when the internal dynamics of chiral molecules are described as an adiabatic evolution in the reduced pseudospin subspace of the two lowest energy levels. Thus, the generalized Stern-Gerlach effect can be used to distinguish molecules with different chiralities, suggesting a discrimination method to separate chiral mixtures.

Model.—We first consider a general case of a symmetry-breaking molecule having a Δ-type or cyclic three-level configuration [e.g., see the left-handed chiral molecule in Fig. 1(a)]. The ground state $|1\rangle$ and the metastable state $|2\rangle$ are coupled to the excited state $|3\rangle$ through spatially varying classical laser fields, with the Rabi frequencies Ω_{13} and Ω_{23}, respectively. In contrast to the Λ-type system, an additional coupling between $|1\rangle$ and $|2\rangle$ is applied by the third classical laser field with the Rabi frequency Ω_{12}.

The total wave function $|\Psi(\mathbf{r})\rangle = \sum_{j=1}^{3} \psi_j(\mathbf{r})|j\rangle$ of the cyclic molecule, where $\mathbf{r}$ denotes the molecular center of mass, is governed by the total Hamiltonian $H = \mathbf{p}^2/(2m) + U(\mathbf{r}) + H_{\rm inn}$, where m is the molecular mass. The trapping potential $U(\mathbf{r}) = \sum_j U_j(\mathbf{r})|j\rangle\langle j|$ is diagonal in the basis of inner states $|j\rangle$, and the inner Hamiltonian $H_{\rm inn}$ contains the free terms $\omega_j|j\rangle\langle j|$ and the Rabi coupling terms $\Omega_{jl}\exp(-i\nu_{jl}t)|j\rangle\langle l| + \mathrm{H.c.}$ ($j = 1, 2, 3$; $l > j$) where ω_j corresponds to the inner level energies (we assume $\hbar = 1$). Here, the frequencies of the three classical optical fields are

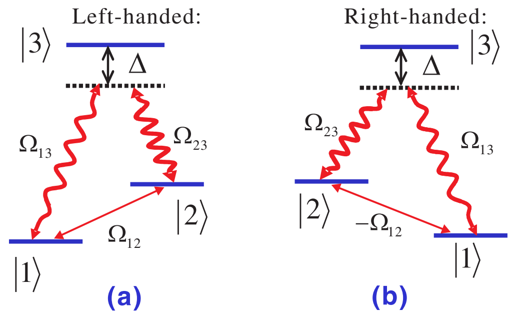

FIG. 1 (color online). Model of three-level Δ-type left-handed (a) and right-handed (b) chiral molecules, coupled to laser beams with Rabi frequencies $\pm\Omega_{12}$, Ω_{13}, and Ω_{23}.

ν_{jl} matching the transition $|j\rangle \rightarrow |l\rangle$ with the Rabi frequencies $\Omega_{jl} = \mu_{jl} E_{jl} = |\Omega_{jl}(t)| \exp(i\phi_{jl})$, respectively; μ_{jl} are the electrical dipole matrix elements, and E_{jl} the envelopes of the electric fields corresponding to the optical fields that couple levels j and l; ϕ_{jl} are the corresponding phases.

We now consider the case that the optical field of Rabi frequency Ω_{12} is resonant to the transition $|1\rangle \rightarrow |2\rangle$, while the other two optical fields are in two-photon resonance with the same single-photon detuning $\Delta = \omega_3 - \omega_2 - \nu_{23} = \omega_3 - \omega_1 - \nu_{13}$ [see Fig. 1(a)]. For position-independent or adiabatically varying Ω_{jl}, the inner Hamiltonian $H_{\rm inn}$ can be rewritten in a time-independent form

$$H'_{\rm inn} = \Delta|3\rangle\langle 3| + \sum_{l>j=1}^{3} \Omega_{jl}|j\rangle\langle l| + \text{H.c.} \tag{1}$$

in the interaction picture.

From now on, we assume large detuning and weak coupling: $|\Delta| \gg |\Omega_{13}| \sim |\Omega_{23}| \gg |\Omega_{12}|$, so that we can use a canonical transformation [14,15] to eliminate the excited level $|3\rangle$ from the Hamiltonian (1). To this end we decompose the inner Hamiltonian as $H'_{\rm inn} = H_0 + H_1 + H_2$ with the zeroth-order Hamiltonian $H_0 = \Delta|3\rangle\langle 3|$, the first-order term $H_1 = \Omega_{13}|1\rangle\langle 3| + \Omega_{23}|2\rangle \times \langle 3|$ + H.c., and second-order term $H_2 = \Omega_{12}|1\rangle\langle 2|$ + H.c. Then the unitary transformation [14,15] $H^{\rm inn}_{\rm eff} = \exp(-S)H'_{\rm inn}\exp(S) \simeq H_0 + [H_1, S]/2 + H_2$ defined by the anti-Hermitian operator $S = (\Omega_{13}|1\rangle\langle 3| + \Omega_{23}|2\rangle\langle 3| - \text{H.c.})/\Delta$ results in the following second-order Hamiltonian:

$$\begin{aligned} H^{\rm inn}_{\rm eff} &= \Delta|3\rangle\langle 3| + \Lambda_1|1\rangle\langle 1| + \Lambda_2|2\rangle\langle 2| \\ &\quad + (g e^{i\Phi}|1\rangle\langle 2| + \text{H.c.}), \end{aligned} \tag{2}$$

where the energy shifts Λ_i are given by $\Lambda_1 = -|\Omega_{13}|^2/\Delta$, $\Lambda_2 = -|\Omega_{23}|^2/\Delta$, and the effective coupling is $g\exp(i\Phi) = \Omega_{12} - \Omega_{13}\Omega^*_{23}/\Delta$. The instantaneous eigenstates of $H^{\rm inn}_{\rm eff}$ are obtained as $|\chi_3\rangle = |3\rangle$, and the dressed states as

$$\begin{aligned} |\chi_1\rangle &= \cos\theta|1\rangle + e^{-i\Phi}\sin\theta|2\rangle, \\ |\chi_2\rangle &= -\sin\theta|1\rangle + e^{-i\Phi}\cos\theta|2\rangle \end{aligned} \tag{3}$$

with the corresponding eigenvalues $\lambda_j = \Lambda_j - (-1)^j g\tan\theta$ for $j = 1, 2$ and $\lambda_3 = \Delta$, where θ is given by $\tan 2\theta = 2g/(\Lambda_1 - \Lambda_2)$.

Induced gauge potentials.—In the new inner dressed-state basis $\{|\chi_1\rangle, |\chi_2\rangle, |\chi_3\rangle\}$, the full quantum state $|\Psi(\mathbf{r})\rangle = \sum_{j=1}^{3}\psi_j(\mathbf{r})|j\rangle$ should be represented as $|\Psi(\mathbf{r})\rangle = \sum_{j=1}^{3}\tilde{\psi}_j(\mathbf{r})|\chi_j\rangle$, where the wave functions $\tilde{\psi} = (\tilde{\psi}_1, \tilde{\psi}_2, \tilde{\psi}_3)^T$ obey the Schrödinger equation $i\partial_t\tilde{\psi} = \tilde{H}\tilde{\psi}$ with the effective Hamiltonian $\tilde{H} = (i\boldsymbol{\nabla} + \underline{\mathbf{A}}(r))^2/(2m) + \underline{V}(\mathbf{r})$ [5,6,16]. Here, the induced gauge potentials, i.e., the vector potential $\underline{\mathbf{A}}(r)$ and the scalar potential $\underline{V}(\mathbf{r})$, are two 3×3 matrices defined by $\mathbf{A}_{j,l} = i\langle\chi_j|\boldsymbol{\nabla}\chi_l\rangle$ and $V_{j,l} = \lambda_j\delta_{j,l} + \langle\chi_j|U(\mathbf{r})|\chi_l\rangle$. The off-diagonal elements of $\underline{\mathbf{A}}$ and $\underline{V}$ can be neglected: the Born-Oppenheimer approximation can be applied to show that they vanish if the adiabatic condition applies [4]. Furthermore, the inner excited state $|\chi_3\rangle = |3\rangle$, whose eigenenergy $\lambda_3 = \Delta$ is much larger than the other inner eigenenergies λ_1 and λ_2, is decoupled from the other inner dressed states. Thus, the three-level cyclic system is reduced to the subsystem spanned by the two lower eigenstates $\{|\chi_1\rangle, |\chi_2\rangle\}$, which are robust to atomic spontaneous emission. This results in an effective spin-1/2 system with pseudospin up and down states $|\uparrow\rangle \equiv |\chi_1\rangle$ and $|\downarrow\rangle \equiv |\chi_2\rangle$.

The Schrödinger equation of the effective two-level system in the pseudospin-1/2 basis $\{|\uparrow\rangle, |\downarrow\rangle\}$ is governed by the diagonal effective Hamiltonian $\tilde{H}_{\rm eff} = H_\uparrow|\uparrow\rangle\langle\uparrow| + H_\downarrow|\downarrow\rangle\langle\downarrow|$, where

$$H_\sigma = \frac{1}{2m}(i\boldsymbol{\nabla} + \mathbf{A}_\sigma)^2 + V_\sigma(\mathbf{r}), \qquad (\sigma = \uparrow, \downarrow). \tag{4}$$

Here, $\mathbf{A}_\sigma = i\langle\chi_\sigma|\boldsymbol{\nabla}\chi_\sigma\rangle$ is the spin-dependent induced vector potential and

$$\begin{aligned} V_\sigma(\mathbf{r}) = \lambda_\sigma + \langle\chi_\sigma|U|\chi_\sigma\rangle + \frac{1}{2m}[\langle\boldsymbol{\nabla}\chi_\sigma|\boldsymbol{\nabla}\chi_\sigma\rangle \\ + |\langle\chi_\sigma|\boldsymbol{\nabla}\chi_\sigma\rangle|^2] \end{aligned} \tag{5}$$

is the reduced optically induced scalar potential [16] for the spin-σ component where $\lambda_{\uparrow,\downarrow} := \lambda_{1,2}$.

We now consider a specific configuration of three Gaussian laser beams copropagating in the $-\hat{z}$ direction. The spatial profiles of the corresponding Rabi frequencies Ω_{jl} are assumed to be of Gaussian form

$$\Omega_{jl} = \Omega^0_{jl} e^{-(x-x_{jl})^2/\sigma^2_{jl}} e^{-ik_{jl}z}, \tag{6}$$

where $j < l = 1, 2, 3$, Ω^0_{jl} are real constants, the wave vectors satisfy $k_{12} + k_{23} - k_{13} = 0$, and the center positions are assumed to be $x_{13} = -x_{23} = \Delta x$, $x_{12} = 0$. The explicit form of the vector potentials are

$$\mathbf{A}_\uparrow = -k_{12}\sin^2\theta\,\hat{\mathbf{e}}_z, \qquad \mathbf{A}_\downarrow = -k_{12}\cos^2\theta\,\hat{\mathbf{e}}_z. \tag{7}$$

Thus, the different spin states of the molecule will have opposite spin-dependent effective magnetic fields $\mathbf{B}_\uparrow = -\mathbf{B}_\downarrow$ according to $\mathbf{B}_\sigma = \boldsymbol{\nabla}\times\mathbf{A}_\sigma$ [6,17].

The internal state of the molecule is prepared in the spin-up and -down states by using the laser beams. The external atomic trap $U(\mathbf{r})$ is turned off at time $t = 0$, and the molecules fall due to gravity with an acceleration G along the direction $\hat{z}$ [18]. The scalar potentials V_σ for spin-up and -down molecules are given explicitly as

$$\begin{aligned} V_\uparrow(\mathbf{r}) &= \lambda_\uparrow + \frac{1}{2m}[k^2_{12}\sin^2\theta(1 + \sin^2\theta) + (\partial_x\theta)^2], \\ V_\downarrow(\mathbf{r}) &= \lambda_\downarrow + \frac{1}{2m}[k^2_{12}\cos^2\theta(1 + \cos^2\theta) + (\partial_x\theta)^2]. \end{aligned} \tag{8}$$

The spin-dependent induced vector potential $\mathbf{A}_\sigma(\mathbf{r})$ and scalar potential $V_\sigma(\mathbf{r})$ lead to the following equations of orbital motion:

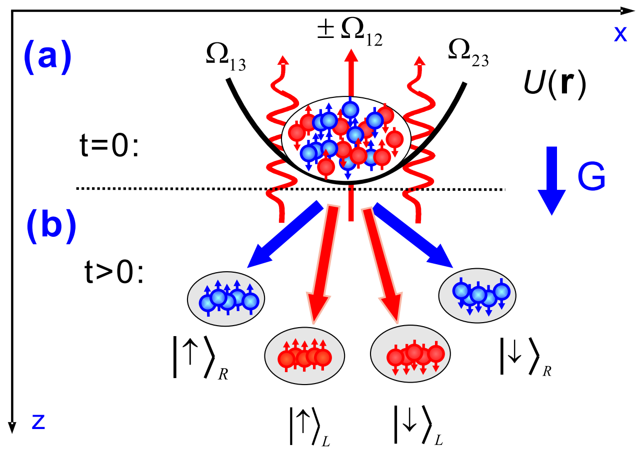

FIG. 2 (color online). Schematic illustration of the generalized Stern-Gerlach experiment of oriented chiral molecules. (a) Mixed chiral molecules trapped by the external potential $U(\mathbf{r})$ are coupled to three laser fields and reduce to the lower dressed internal state space of spin-up and spin-down. (b) After the trap potential is switched off at time $t = 0$, the molecules will fall under gravity $G = 9.8\ \mathrm{m/s^2}$. Because of the chirality-dependent induced gauge field, molecules with different chirality will experience different Stern-Gerlach effects.

$$\dot{x}_\sigma = \frac{p_{\sigma,x}}{m}, \qquad \dot{z}_\sigma = \frac{p_{\sigma,z} - A_{\sigma,z}}{m}, \qquad \dot{p}_{\sigma,z} = mG,$$
$$\dot{p}_{\sigma,x} = \frac{1}{m}[(\partial_x A_{\sigma,z})p_{\sigma,z} - A_{\sigma,z}\partial_x A_{\sigma,z}] - \partial_x V_\sigma. \tag{9}$$

Hence, there will be a Stern-Gerlach-like effect, i.e., different spatial motion of the cyclic molecules corresponding to different initial states of spin up and down. In contrast to the standard Stern-Gerlach effect, the effective magnetic field is not required to have a gradient. Here and in the following, we treat the orbital motion as classical because of the large molecular mass and weak effective gauge potentials.

Generalized Stern-Gerlach effect.—In the large detuning and weak-coupling limit, the above approach works well for any type of cyclic three-level system. It can also be applied in an experimentally feasible scheme to detect the chirality of molecules, since left- and right-handed molecules have different Stern-Gerlach-like effects. Physically, left- and right-handed molecules have the same intrinsic properties except the antisymmetry of the total phase for the three coupled Rabi frequencies [9,10]. Hence, we can define $\Omega^L_{ij} \equiv \Omega_{ij}$ as the Rabi frequencies for the left-handed molecules, and define the Rabi frequencies for the right-handed ones: $\Omega^R_{12} \equiv -\Omega_{12}$ and $\Omega^R_{13} \equiv \Omega_{13}$, $\Omega^R_{23} \equiv \Omega_{23}$ for the same coupling optical fields [see Figs. 1(a) and 1(b)]. Therefore the difference in chirality leads to two different effective couplings,

$$g_{L/R}e^{i\Phi_{L/R}} = \pm\Omega_{12} - \frac{1}{\Delta}\Omega_{13}\Omega^*_{23} \tag{10}$$

(the index L on the left-hand side corresponds to the positive sign on the right-hand side) which results in two different effective inner Hamiltonians

$$H^{\mathrm{inn}(Q)}_{\mathrm{eff}} = \Delta|3\rangle_{QQ}\langle 3| + \Lambda_1|1\rangle_{QQ}\langle 1| + \Lambda_2|2\rangle_{QQ}\langle 2| + (g_Q e^{i\Phi_Q}|1\rangle_{QQ}\langle 2| + \mathrm{H.c.}), \qquad (Q = L, R). \tag{11}$$

Initially, the mixed left- and right-handed oriented molecules, which are spatially confined due to the external trap potential, are subject to three coupling optical fields as seen in Fig. 2(a) and reduced to the spin-state space $\{|\uparrow\rangle_{L/R}, |\downarrow\rangle_{L/R}\}$. At time $t = 0$, the external trap potential is turned off and the molecules will fall due to gravity. As in the above consideration for the general case of cyclic-type molecules in Eqs. (2)–(9), we obtain optically induced potentials and different molecular classical trajectories for left- and right-handed molecules, respectively. This is schematically illustrated in Fig. 2(b), which shows that the generalized Stern-Gerlach effect splits the initial cloud into four subsets, since the effective gauge potentials depend both on spin and chirality.

To make this picture of a generalized Stern-Gerlach effect more quantitative, we show in Fig. 3 the typical

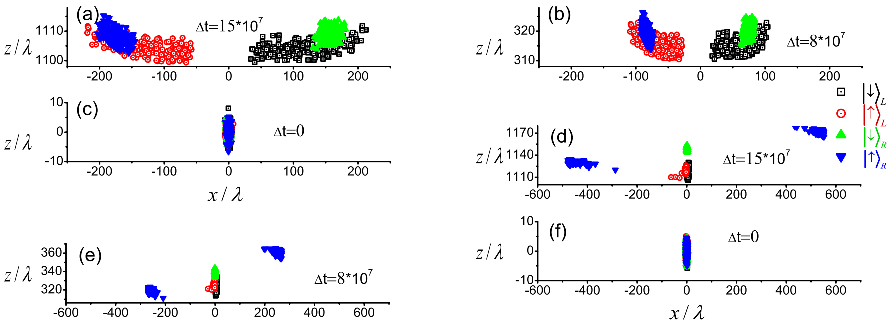

FIG. 3 (color online). The positions of an oriented molecular ensemble with an initial Gaussian position distribution $[\rho(x, z) = (2\pi\sigma_r^2)^{-1}\exp[-(x^2 + z^2)/\sigma_r^2]$ with $\sigma_r = 3\lambda]$ at different times. The positions x, z are in units of λ (the wavelength for the lower transition $\lambda = 2\pi/k_{12} = 2\pi c/\nu_{12}$ with c the optical velocity in vacuum; typically, $\lambda \sim 1\ \mu\mathrm{m}$). The following parameters are used: (a)–(c): $\Omega^0_{12} = \Omega^0_{13}\Omega^0_{23}/\Delta \sim 10^{-6}\Delta$; (d)–(f): $\Omega^0_{12} = \Omega^0_{13}\Omega^0_{23}/\Delta \sim 10^{-4}\Delta$ (the detuning $\Delta \sim 10^{10}$ Hz is large). Here, we assume $\sigma_{13} \equiv \sigma_{23} = 10\lambda$, $\sigma_{12} = 7\lambda$, $\Delta x = 3\lambda$, $\Omega^0_{13} \equiv \Omega^0_{23}$. The molecular mass is taken to be 100 times the proton mass.

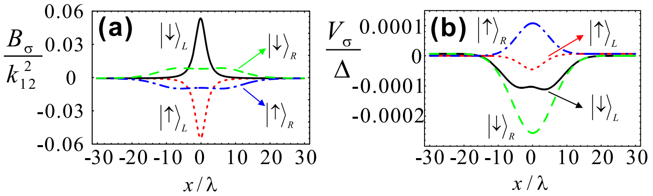

FIG. 4 (color online). (a) Effective magnetic field corresponding to Fig. 3 (in units of $k_{12}^2 \equiv 4\pi^2/\lambda^2$); (b) Effective scalar potential corresponding to Figs. 3(d)–3(f).

position of an oriented ensemble of mixed left- and right-handed molecules and spin states subject to gravity (in the $\hat{z}$ direction). For temperatures below 1 μK, the initial velocity of the molecules can be neglected. Figures 3(a)–3(c) show the $\hat{x}$-$\hat{z}$-plane positions of such a molecular ensemble with an initial Gaussian position distribution at the origin at different times. The spatial separation of molecules with different spin projections is clearly visible. By choosing a different value of Ω_{12}^0 and $\Omega_{13}^0\Omega_{23}^0/\Delta$, we also obtain a spatial separation of molecules with different chirality; see Figs. 3(d)–3(f). The separation is partial in the following sense: for our choice of parameters, right-handed molecules in the spin-up state are deflected to finite values of x, whereas the other three components are not deflected and their trajectories remain close to $x = 0$. By changing $\Omega_{12}^0 \rightarrow -\Omega_{12}^0$, the role of left and right in Figs. 3(a)–3(f) is interchanged. In Fig. 4 we show the effective magnetic fields (i.e., the curl of the vector potentials) and scalar potentials leading to this behavior. Figure 4(a) shows the effective magnetic fields corresponding to all the subplots in Fig. 3 [the effective magnetic field is the same in Figs. 3(a)–3(f)]. For the situation in Figs. 3(a)–3(c), the effects of the scalar potential (which is not shown) can be neglected: the magnetic fields are dominant and make the molecules in the spin-up state move along the $-x$ direction (spin-down states along the x direction). In Figs. 3(d)–3(f) the scalar potentials are dominant [Fig. 4(b)] and will trap the molecules in the area around $x = 0$, except for the $|\uparrow\rangle_R$ molecules that are deflected.

Stern-Gerlach experiments can also be used to obtain and measure superpositions of spin states. However, our effect described above does not work for superpositions of left- and right-handed chiral states (even if many studies about teleporting, preparing, and measuring superpositions of chiral states [19] appeared recently), since this would require considering higher excited symmetric or antisymmetric states. We will leave this interesting question for future works.

Although the protocol presented here is idealized since intermolecular interactions are neglected, it provides a promising way to spatially separate molecules of different chiralities. A similar generalized Stern-Gerlach effect has been proposed for Λ-type systems where the Rabi frequencies Ω_{12} between the two lower inner states vanish [6]. However, this effect is chirality independent. Thus, in contrast to our configuration, the effect discussed in [6] cannot be used to distinguish and separate left- and right-handed molecules.

Conclusion.—In conclusion, we have studied orbital effects of internal adiabatic transitions on the center-of-mass motion of oriented chiral molecules. We have shown that under the conditions described above, the center-of-mass motion of the molecules depends on both chirality and spin due to optically induced gauge potentials and can be interpreted as a generalized Stern-Gerlach effect. This leads to the possibility of spatially separating molecules of different chiralities.

This work was supported by the European Union under Contract No. IST-3-015708-IP EuroSQIP, by the Swiss NSF, and the NCCR Nanoscience, and also by the NSFC and NFRPC of China.

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