First-principles calculations of steady-state voltage-controlled magnetism: Application to x-ray absorption spectroscopy experiments

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Recent x-ray absorption experiments have demonstrated the possibility to accurately monitor the magnetism of metallic heterostructures controlled via a time-independent perturbation caused, for example, by a static electric field. Using a first-principles, nonequilibrium Green's function scheme, we show how the measured dichroic signal for the corresponding steady-state situation can be related to the underlying electronic structure and its response to the external stimulus. The suggested approach works from the infinitesimal limit of linear response to the regime of strong electric field effects, which is realized in present experimental high-sensitivity investigations.

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The creation, control, and detection of spin-polarized currents lie at the heart of spintronics. Accordingly, a number of magneto-optical experiments have been proposed over the years to explore related magnetic phenomena and corresponding materials. Obviously, x-ray-based measurement techniques seem to be especially suited for this purpose because they provide a high signal-to-noise ratio, precise targeting of individual chemical species, and tunable sensitivity to either the surface or bulklike properties of a sample, depending on the photon energy [1,2].

In particular, the time dependence of magnetic properties has been studied over a broad range of timescales by means of magneto-optical techniques, addressing for example ultrafast demagnetization [3–9] or optical manipulation of the magnetic order [10–13]. In general, the most comprehensive information can be acquired by corresponding experiments using pump-probe techniques. Early experiments in the field, such as the time-dependent magneto-optical Kerr effect (MOKE) or x-ray magnetic circular dichroism (XMCD) experiments [3,4], have been performed by controlling the investigated system via an external magnetic field and using different delay times for the read-out via x-ray pulses.

Another type of interesting experiments exploits the XMCD to study the impact of an external static electric field on the magnetization. For the resulting out-of-equilibrium situation one may have pure charge rearrangements with a continuous flow of electric charge prevented by applying the electric field across an interface to vacuum or an insulating

layer, i.e., having a capacitorlike experimental setup [14,15]. For the combination of conducting subsystems, on the other hand, a steady-state out-of-equilibrium situation will be created with a constant electric current flowing [16,17]. In this case one may focus on the electric-field-induced change of the magnetization longitudinal [16] or transverse [17] with respect to the electric field. The electric-field-induced electric current will in general be accompanied by a spin current that might be used, for example, for switching the magnetization via the spin transfer torque (STT), spin-orbit torque (SOT), or the spin Hall effect (SHE) [18]. Accordingly, the question concerning the connection between the observed XMCD and the induced spin current arises in a natural way.

In the following, a theoretical description of the electric-field-induced XMCD for the case of a conducting system is given, with a focus on the longitudinal setup investigated by Kukreja *et al.* [16]. These authors investigated the bilayer system Co/Cu by XMCD measurements at the Cu L_3 edge. It was found that a voltage applied across the layer system changes the XMCD spectra primarily in the vicinity of the Fermi level. By switching the sign of the voltage it was possible to separate in a reliable way the electric-field-induced contribution from that due to the so-called proximity effect (see below). This allowed in particular to demonstrate the linear dependence of the induced changes for the XMCD spectra on the applied voltage. Here, we present an *ab initio* description of the observed phenomenon, that accounts in particular for the out-of-equilibrium situation when a finite voltage is applied to a conducting system. To deal with the XMCD in this case, an appropriate expression for the x-ray absorption coefficient together with a corresponding extension of the XMCD sum rules [19–22] are suggested. This allows us to relate the XMCD spectra of an atom to its spin and orbital moments under out-of-equilibrium conditions.

Considering the electronic structure of a system out of equilibrium, the occupied and unoccupied states can be represented in an appropriate way in terms of the lesser and

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FIG. 1. Sketch of the layered system considered: A Co/Pd layer system is connected to the left and to the right to Cu leads. The Co and Pd layers, together with a few neighboring Cu layers, form the central so-called interaction zone.

greater Green's functions, respectively [23]. Here, we deal with the steady-state situation encountered for a layered system exposed to a constant electric field across the layers, or equivalently a layered system connected to the left and right leads with a corresponding voltage drop Φ in between. In this case, it is most convenient to consider the lesser and greater Green's functions, $G_{\Phi}^{<}(E)$ and $G_{\Phi}^{>}(E)$, respectively, as functions of the energy E and dependent on the applied voltage Φ . For the setup of a layered system as sketched schematically in Fig. 1, it was shown by several authors [24–26] that the Green's functions $G_{\Phi}^{<}(E)$ can be calculated by means of a Dyson-like equation, that relates the various spatial regions and Green's functions, through an appropriately defined self-energy $\Sigma_{\Phi}^{<}(E)$. This way one gets $G_{\Phi}^{<}(E)$ by the expression [27–29]

$$G_{\Phi}^{<}(E) = G_{\Phi}^r(E) \Sigma_{\Phi}^{<}(E) G_{\Phi}^a(E) \quad (1)$$

in terms of the retarded and advanced Green's functions, $G_{\Phi}^r(E)$ and $G_{\Phi}^a(E)$, respectively, that in turn can be calculated using standard techniques [30]. In this context, the finite voltage Φ determines the finite and fixed difference in the chemical potentials of the left and right leads, $\mu_L = E_F + \Phi/2$ and $\mu_R = E_F - \Phi/2$, respectively, where E_F is the Fermi level of the nonbiased, i.e., unperturbed reference system with $\Phi = 0$ V. To deal with the gradual voltage drop across the region between the leads, denoted the interaction zone in Fig. 1, a corresponding model suggested in the literature [31,32] was adopted. (See Supplemental Material SM1 [33] for more details.) The nonequilibrium Green's function formalism we employ is nonperturbative, meaning that the voltage Φ does not have to be small.

The scheme to calculate the lesser and greater Green's functions, briefly sketched above, was implemented within

the framework of local spin density approximation making use of the Korringa-Kohn-Rostoker (KKR) Green's function technique as suggested by Ogura and Akai [34]. As the study aimed among others at the calculation of spin-orbit-induced XMCD spectra, a fully relativistic formulation of the scheme was used on the basis of the four-component Dirac equation, i.e., all Green's functions are 4×4 matrix functions [35].

With the lesser and greater Green's functions $G_{\Phi}^{<}(E)$ available, many electronic properties of interest can be calculated straightforwardly. For example, the density of states (DOS) related to the occupied ($<$) and unoccupied ($>$) states is given by $n_{\Phi}^{<(d)}(E) = -1/\pi \text{Trace } G_{\Phi}^{<(d)}(E)$, where the angular-momentum representation used for the Green's function [35] allows one in a simple way to project out the contribution due to d electrons as indicated by the optional index d . In a similar manner, one has for the spin magnetic moment $m_{\Phi}^{(d)}$ associated with the occupied states

$$m_{\Phi}^{(d)} = -\frac{1}{\pi} \int_{-\infty}^{E_F + |\Phi|/2} dE \text{Trace } \beta \sigma_z G_{\Phi}^{<(d)}(E), \quad (2)$$

where β is one of the standard Dirac matrices, σ_z a 2×2 Pauli matrix [36], and the integration regime has been restricted according to the maximum of μ_L and μ_R . (See Supplemental Material SM2 [33] for more details.)

For the steady-state situation considered here, an appropriate expression for the x-ray absorption coefficient $\mu_{\Phi}^{\vec{q},\lambda}(\omega)$ can straightforwardly be derived by replacing in the standard expression for an unperturbed ground state [37] the product of the retarded Green's function and the Fermi distribution at a given temperature, i.e., $G^r(E)[1 - f_T(E)]$, by the greater Green's function $G_{\Phi}^{>}(E)$ representing the unoccupied final states. This leads to

$$\mu_{\Phi}^{\vec{q},\lambda}(\omega) \propto \sum_{i \in \text{occ}} \langle \Psi_i | X_{\vec{q},\lambda}^{\dagger} G_{\Phi}^{>}(E_i + \omega) X_{\vec{q},\lambda} | \Psi_i \rangle, \quad (3)$$

where Ψ_i represents an occupied initial core state i while $X_{\vec{q},\lambda}$ stands for the interaction of the electrons with the photons with wave vector $\vec{q}$, energy ω , and polarization λ [37]. Dealing with $X_{\vec{q},\lambda}$ we make use of the dipole approximation. Accordingly, the index $\vec{q}$ can be omitted in the following. Finally, for the case $\Phi = 0$ V, the rather general expression for $\mu_{\Phi}^{\vec{q},\lambda}(\omega)$ in Eq. (3) is of course fully equivalent to the standard one in terms of $G^r(E)$ [37]. This is demonstrated by a numerical example in the Supplemental Material SM3 [33].

An extremely attractive tool to interpret experimental as well as theoretical XMCD data, i.e., the difference in absorption of left or right circularly polarized x rays, is provided by the XMCD sum rules [19–22]. Focusing on $L_{2,3}$ spectra, the spin-related sum rule is given by

$$\frac{m^d + 7T_z^d}{N_h^d} = \frac{\int_{E_{\min}}^{E_{\text{cutoff}}} [\mu_{L_3}^{\lambda=+}(\omega) - \mu_{L_3}^{\lambda=-}(\omega)] d\omega - 2 \int_{E_{\min}}^{E_{\text{cutoff}}} [\mu_{L_2}^{\lambda=+}(\omega) - \mu_{L_2}^{\lambda=-}(\omega)] d\omega}{\int_{E_{\min}}^{E_{\text{cutoff}}} [\bar{\mu}_{L_3}(\omega) + \bar{\mu}_{L_2}(\omega)] d\omega}, \quad (4)$$

where m^d is the spin magnetic moment associated with d states, T_z^d denotes the magnetic dipole or asphericity

term, $N_h^d = 10 - \int_{-\infty}^{E_F} n^d(E) dE$ is the number of d -like holes derived from the d -like DOS $n^d(E)$ below the Fermi

level E_F , and $\bar{\mu}_{L_{2(3)}}(\omega)$ stands for the polarization averaged absorption coefficient for the L_2 (L_3) edge. All energy integrals in Eq. (4) run up to the cutoff energy E_{cutoff} for which the integrated d -like DOS covers ten d states, i.e., $\int_{-\infty}^{E_{\text{cutoff}}} n^d(E) dE = 10$.

The sum rule in Eq. (4) was initially derived under certain assumptions [22,37] for the situation that no external voltage is applied to the system. It is not *a priori* clear to what extent these assumptions remain valid if $\Phi \neq 0$. Nevertheless, Eq. (4) can be straightforwardly adapted to the situation of a finite applied voltage Φ by calculating the absorption coefficients $\mu_{\Phi}^{\lambda}(\omega)$ by means of Eq. (3) and using the greater Green's function $G_{\Phi}^{>}(E)$. The quantities m^d , T_z^d , and N_h^d , on the other hand, have to be calculated accordingly using the lesser Green's function $G_{\Phi}^{<}(E)$. Finding the number of d holes N_h^d requires in particular an integration of the DOS $n_{\Phi}^{<d}(E)$ related to $G_{\Phi}^{<}(E)$ up to the energy where it goes to zero. This upper threshold energy lies typically above the original Fermi energy E_F for the case $\Phi = 0$ V. On the other hand, the lower threshold energy from which unoccupied states may contribute to the x-ray absorption via $G_{\Phi}^{>}(E)$ is given by $E_{\text{min}} \leq E_F - |\frac{\Phi}{2}|$.

The experiment of Kukreja *et al.* [16] was performed on Co/Cu, involving changes of the moments at Cu of order $10^{-3} \mu_B$. Achieving such a numerical accuracy for the present out-of-equilibrium steady-state situation would be extremely demanding. To avoid numerical problems and to ensure pronounced field-induced changes of the magnetic properties suitable for further analysis, the bilayer system Co/Pd has been considered in this work, as Pd has a much higher spin polarizability than Cu. Otherwise, the setup sketched in Fig. 1 followed essentially the work of Kukreja *et al.* [16] on Co/Cu, i.e., we investigated a fcc (001) textured Co/Pd bilayer system consisting of eight layers of Co with their magnetization oriented perpendicular to the layers (out of plane) and eight layers of Pd. On both sides four buffer layers of Cu have been added to allow for a smooth connection to the fixed Cu leads with their respective chemical potential shifted by the voltage drop Φ .

Figure 2(a) shows the resulting profile of the spin magnetic moment m_{Φ}^n for the Co/Pd bilayer for $\Phi = 0$ and ± 0.34 V, where n is the layer index. For $\Phi = 0$ V one finds an enhancement for the Co moments at the Co/Pd interface [38], while for the Pd layers there is an appreciable induced moment that decays rapidly with the distance from the interface (proximity effect). These well-known features of the magnetization profile at the Co/Pd interface are obviously essentially unchanged when a finite voltage Φ is applied, that modifies the magnetic moments depending on the sign of Φ . The corresponding electric-field-induced contribution to the magnetization profile $\Delta m_{\Phi}^n = m_{\Phi}^n - m_0^n$ is shown for $\Phi = +0.34$ V in Fig. 2(b). As one can see, the changes of the Co and Pd moments are of the same order of magnitude but interestingly of different sign. In addition, one notes that Δm_{Φ}^n has nearly exclusively to be ascribed to the part of the d electrons ($\Delta m_{\Phi}^{d,n}$). The layer-resolved induced magnetic moments, and with these also the averaged induced moments of Co and Pd, scale fairly well linearly with the applied voltage Φ (see below).

FIG. 2. (a) Layer- (n -) resolved profile of the spin magnetic moment m_{Φ}^n (in μ_B) for the investigated Co/Pd bilayer system for three applied voltages: $\Phi = -0.34$ (red bars), 0 (black bars), and $+0.34$ V (blue bars). (b) Electric-field-induced contribution to the profile $\Delta m_{\Phi}^n = m_{\Phi}^n - m_0^n$ (solid bars) for the voltage drop $\Phi = +0.34$ V with $\Delta m_{\Phi}^{d,n}$ its part due to the d electrons (shaded bars).

These observed properties of the induced magnetic moments suggest an alternative description of the phenomenon of an electric-field-induced magnetization by means of Kubo's linear response theory, i.e., within a perturbative approach as opposed to the nonperturbative nonequilibrium Green's function formalism. The corresponding Kubo-Bastin-like expression for the nonlocal response coefficient p_{zz}^{nm} that gives for layer n the induced magnetization along the z axis due to an electric field in layer n' along the same direction can be found in the Supplemental Material SM4 [33]. Corresponding numerical results for the local layer-dependent polarization coefficient p_{zz}^n defined by the sum $\sum_{n'} p_{zz}^{nn'}$ are given in Fig. 3 for the investigated Co/Pd bilayer. Obviously, the profile of p_{zz}^n is in full accordance with the profile of the induced magnetic moment Δm_{Φ}^n shown in Fig. 2(b). In particular, the different sign for the induced moment for Co and Pd is fully confirmed by the linear response calculations. In addition, we find also that p_{zz}^n is by far dominated by its part stemming from the d electrons ($p_{zz}^{d,n}$). Here, it should be stressed that a perfect one-to-one correspondence of $\Delta m_{\Phi}^{(d)n}$ in Fig. 2(b) and $p_{zz}^{(d)n}$ in Fig. 3, respectively, cannot be expected as the evaluation of the induced magnetic moment requires the nonlocal response coefficient $p_{zz}^{nn'}$ together with the local electric field $E_z^{n'}$ for the various layers. However, the latter one will depend in a nontrivial way on the layer index n' as it is suggested by the local electrical conductivity σ_{zz}^n that varies in an appreciable way from layer to layer for the Pd subsystem as can be seen

FIG. 3. Layer- n -resolved Edelstein coefficient p_{zz}^n (solid bars) together with its part $p_{zz}^{d,n}$ due to the d electrons (shaded bars). In addition, the layer-resolved charge conductivity σ_{zz}^n (orange circles) is shown.

in Fig. 3 (orange circles). Nevertheless, the comparison of the profiles of $\Delta m_{\Phi}^{(d)n}$ and $p_{zz}^{(d)n}$ in Figs. 2(b) and 3, respectively, clearly shows that the electric-field-induced magnetization is first of all a manifestation of the Edelstein effect [39,40]. Concerning this, it should be stressed that the lack of inversion symmetry is the central precondition for the occurrence of the Edelstein effect [39]. This is of course given for the Co/Pd interface region.

The proximity effect mentioned above, i.e., the occurrence of an induced spin magnetic magnetization in a nonmagnetic metal at the interface to a ferromagnetic metal, could be demonstrated in the past for several systems by means of XMCD measurements on the nonmagnetic metal. Prominent examples for this are Pt in Co/Pt [41] but also Cu in Co/Cu [42] with the high and small, respectively, spin susceptibility of the nonmagnetic metal reflected by the correspondingly high and small induced spin magnetic moments that differ by one to two orders of magnitude (see also Fig. 2).

In an earlier study we predicted that the magnetization induced by an external magnetic field should give rise to a corresponding XMCD signal that scales in a one-to-one manner with the spin and orbital susceptibilities [43]. This could indeed be demonstrated experimentally for the nonmagnetic metals Pt and Pd [44] but also for Au [45]. In a completely analogous way, one can expect that the magnetization induced by an external electric field via the Edelstein effect leads also to the occurrence of an XMCD in the case of an otherwise nonmagnetic system. For an element in a system with a spontaneous or induced magnetization, on the other hand, this mechanism will alter the already present XMCD spectrum accordingly. This was indeed found when calculating the Pd $L_{2,3}$ spectra for the investigated Co/Pd bilayer system when a finite-voltage drop Φ is applied (see Supplemental Material SM5 [33]). To deduce the relationship of the additional XMCD signal and spin magnetic moments induced by the electric field, the $L_{2,3}$ XMCD spectra for the individual Pd layers have been calculated in a first step for the voltages $\pm\Phi$. Taking for each layer the difference leads accordingly to the field-induced contribution of the XMCD spectra, that have

FIG. 4. Electric-field-induced change in the spin magnetic moment of the d states of Pd, as a function of the applied voltage Φ , averaged over the Pd layers: $\langle \Delta m_{\Phi}^d \rangle$ calculated directly from the lesser Green's function $G_{\Phi}^{<}(E)$ via Eq. (2) (open diamonds) and $\langle \Delta m_{\Phi}^{\text{XMCD}d} \rangle$ deduced from the XMCD spectra at the $L_{2,3}$ edges via the modified sum rules [see Eq. (4)] (solid circles).

been analyzed by making use of the modified version of the spin sum rule given in Eq. (4). This leads finally to the field-induced changes of the spin magnetic moments $\Delta m_{\Phi}^{\text{XMCD}d}$ of the d electrons as deduced from the XMCD spectra. As a corresponding experiment does not allow one to distinguish the XMCD spectra of the individual Pd layers n , the average over all these have been taken. This average value $\langle \Delta m_{\Phi}^{\text{XMCD}d} \rangle$ is given in Fig. 4 as a function of the applied voltage Φ with the corresponding averaged spin magnetic moment $\langle \Delta m_{\Phi}^d \rangle$ calculated directly from the lesser Green's function $G^{<}(E)$ via Eq. (2). Keeping in mind the various approximations used to derive the XMCD sum rules [37] the agreement between the directly and XMCD-derived moments is rather satisfying. This finding is very similar to the results of studies on systems with the magnetization occurring spontaneously or induced by the proximity effect [46].

The close connection between the change in the spin magnetic moment $\langle \Delta m_{\Phi}^d \rangle$ calculated directly for the Pd layers and $\langle \Delta m_{\Phi}^{\text{XMCD}d} \rangle$ deduced from the spectroscopic data clearly shows that the electric-field-induced XMCD indeed reflects in a one-to-one manner the induced spin magnetic moments for a steady-state situation. Accordingly, we conclude that the experimental findings of Kukreja *et al.* [16] for Co/Cu have to be interpreted as well this way, i.e., that the observed additional XMCD signal due to the applied voltage has to be seen as a rather direct measure for the electric-field-induced spin magnetic moment and as a manifestation of the Edelstein effect. The fact that the calculated as well as the observed field-induced changes of the XMCD spectra occur primarily in the vicinity of the Fermi level supports this conclusion and interpretation of the experimental findings. Nevertheless, it should be mentioned that the observed XMCD spectra may have additional contributions due to spin accumulation caused by the specific features of the experimental setup not accounted for within the present theoretical study.

In summary, a numerical study on the electric-field-induced changes in the electronic, magnetic, and spectroscopic properties of the ferromagnet/nonmagnet bilayer system Co/Pd has been presented. The field-induced magnetic moments were found to scale essentially linearly with the applied voltage. This finding as well as additional linear response calculations allow us to ascribe the induced magnetic moments to the Edelstein effect. To make contact with recent XMCD investigations on Co/Cu an appropriate expression for the calculation of the x-ray absorption coefficient for a steady-state out-of-equilibrium situation was suggested. Together with a corresponding extension of the XMCD sum rules, the one-to-one corresponding relation between the electric-field-induced contributions to the spin magnetic moment and

XMCD spectra could be demonstrated. From this it could be concluded that the results of a recent experimental XMCD study on the bilayer system Co/Cu first of all reflect the field-induced magnetic moment in the vicinity of the Co/Cu interface.

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